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Short communication

Electrode surface nanostructuring via nanoparticle electronucleation for signal enhancement in electrochemical genosensors

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ABSTRACT

Biosensor read out signals can be enhanced by carefully designing the transducer surfaces to achieve an optimal interaction between the recognition elements immobilised and the targeted analyte. This is particularly evident in the case of genosensors, where spacing and orientation of immobilised DNA capture probes need to be controlled to maximise subsequent surface hybridisation with the target sequence and achieve high binding signals. Addressing this goal, we present a novel approach based on the surface nanostructuring of glassy carbon electrodes (GCEs) towards the development of highly sensitive electrochemical genosensors. Gold nanoparticles were sequentially electrochemically nucleated on glassy carbon electrodes to form dense arrays of randomly distributed gold nanodomains. The number density of the electronucleated nanoparticles could be increased by repeatedly alternating between a short electronucleation step and the subsequent insulation of the nucleated nanoparticles with thiolated DNA probes. This approach allowed for the creation of highly structured surfaces whilst preventing aggregation of nanoparticles. The performances of planar gold electrodes and that of the nanopatterned surfaces prepared following several rounds of deposition were compared for the amperometric detection of DNA. Three rounds of deposition exhibited the highest sensitivity (44.89 nA × nM^{-1}), with a dynamic detection range spanning from 0.53 nM to 25 nM of the targeted sequence, i.e. one order of magnitude lower than that obtained for the planar gold electrodes. The use of the nanostructured surface we report here may find application not only in DNA biosensors but also for any sensing application requiring highly sensitive measurements.

1. Introduction

Electrochemical DNA biosensors have been intensively investigated due to their potential for rapid and low cost DNA testing. Some of the applications of these sensors include gene expression monitoring (Berney et al., 2000), diagnosis of genetic disorders (Alfonta et al., 2001; Patolsky et al., 2003), investigation of DNA damage (Oliveira Brett et al., 2007), bio-analysis of environmental pollution (Bagni et al., 2006) as well as food quality control and traceability (Bettazzi et al., 2008; Marmiroli et al., 2008).

The performance of DNA biosensors is dependent on the overall efficiency of the surface hybridisation event. Short DNA probes are commonly immobilised onto metal surfaces via the introduction of a thiol moiety at the 5′-end of the probe sequence to enable the formation of highly packed DNA self-assembled monolayers (SAMs). In order to optimise the hybridisation process, parameters such as the surface density and orientation of the immobilised DNA probes require accurate control, as several reports have demonstrated that the rate of surface hybridisation decreases with increasing DNA probe density (Henry et al., 1999; Peterson et al., 2001; Sanchez-Pomales et al., 2007; Tokuhisa et al., 2009; Wilkins Stevens et al., 1999; Yguerabide and Ceballos, 1995). On the other hand, although a low DNA probe density could solve steric problems whilst the probe orientation can be kept optimum, the electrochemical signal generated may be too low to be of any analytical value.

Controlling the surface density of the probes continues to be a challenge that needs to be addressed and both top-down and bottom-up fabrication approaches have been described for the preparation of DNA-modified substrates of controlled surface probe density. In particular, backfilling (Berganza et al., 2007; Levicky et al., 1998) and co-immobilisation (Henry et al., 2010; Steel et al., 2000; Tokuhisa et al., 2009) approaches have been widely reported. The former strategy consists in the initial chemisorption of thiolated DNA onto the metal surface. This step leads to the formation of poorly organised monolayer and surfaces are subsequently

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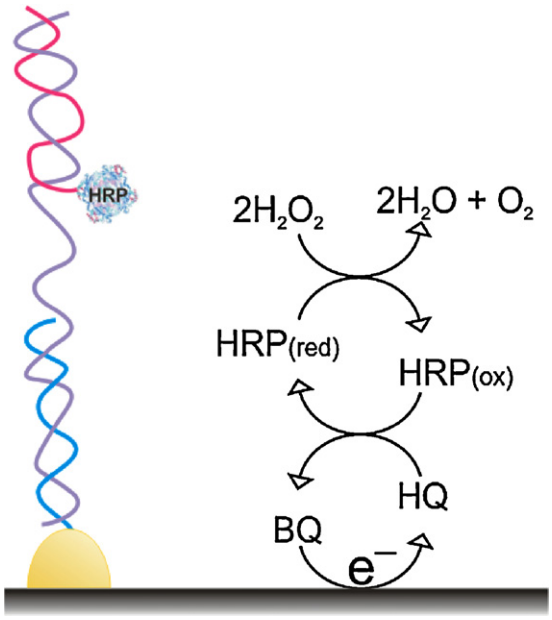

Fig. 1. The electrochemical sandwich assay employed for the detection of LTA and the corresponding electron-relay for generation of chronoamperometric signal. (HQ hydroquinone, BQ benzoquinone, HRP horseradish peroxidase, H_2O_2 hydrogen peroxide, O_2 oxygen.)

backfilled with a second shorter alkanethiol in order to complete the monolayer as well as orientating the DNA strands away from the surface. Even though this technique is the most commonly used approach in electrochemical DNA sensors (Herne and Tarlov, 1997), a problem of surface reproducibility remains (Berggren et al., 2001). In contrast, the co-immobilisation strategy consists in a single step modification where thiolated DNA and short-chain alkanethiols are pre-mixed in a given ratio, which translates into the formation of well-defined monolayers at the metal surface and consequently better sensor-to-sensor reproducibility (Henry et al., 2010; Steel et al., 2000; Tokuhisa et al., 2009). Other reported strategies to address DNA surface density consist in the selective electrochemical desorption of DNA or sacrificial alkanethiol SAMs (Henry et al., 2009; Sanchez-Pomales et al., 2007; Satjapipat et al., 2001), the initial immobilisation of DNA probe–target sequence duplexes followed by thermal denaturation (Peterson et al., 2001), the dendron-controlled spacing of probes (Choi et al., 2004; Oh et al., 2005) or the direct grafting of probe sequences at surfaces (Liu and Xu, 1997; Liu and Liu, 2005).

Direct transducer surface nano-structuring presents an excellent potential for the optimal spacing of DNA probes at the transducer surface whilst at the same time enhancing transducer sensitivity. Metallic nanostructures can be used to define nanoscale immobilisation domains, where a few DNA strands are immobilised at a time, thus limiting packing and consequently steric hindrance and electrostatic repulsion during hybridisation, as well as enhancing the signal due to the unique catalytic properties present at the nanoscale. Surface nanostructures can be realised in a variety of ways, including the random electrochemical nucleation and deposition of metal nanoparticles on an electrode substrate for electrochemical genosensors (Yang et al., 2007), as well as the controlled positioning of metallic nanoparticles using scanning probe microscopy (SPM) techniques (Hugelmann et al., 2005; Leiva, 2006) or nanolithography (Demers et al., 2002).

The work reported here presents a novel approach to the fabrication of electrochemical DNA biosensors based on the formation of DNA SAMs on electrochemically nucleated gold nanoparticles at the surface of glassy carbon electrodes (GCEs). We found that electrochemical nucleation of gold lead to the formation of highly active gold nanoparticles, the density number of which could be tightly controlled by performing sequential nucleation rounds. Following one round of deposition, the freshly deposited nanoparticles were capped with thiolated DNA probes and mercaptoethanol before being exposed to a second round of deposition. This approach allowed us to progressively increase the number density of the nanoparticles whilst preventing aggregation and the formation of a full gold layer. Using a model system for the detection of the lymphotoxin-α gene (LTA) in a "sandwich" format with an enzyme labeled reporting probe, the sensitivity of biosensors prepared after 1–4 rounds of deposition was compared and offered markedly lower detection limits.

2. Materials and methods

2.1. Reagents

Potassium tetrachloro aurate (III) ($KAuCl_4$, 99.995%, Aldrich), sulphuric acid (H_2SO_4, 95%, Scharlau), potassium nitrate (KNO_3, 99%, Sigma), sodium perchlorate ($NaClO_4$, 98%, Sigma), sodium chloride (NaCl, synthesis grade, Scharlau), potassium dihydrogenphosphate (KH_2PO_4, Fluka), hydroquinone ($C_6H_4(OH)_2$, 99%, Sigma), hydrogen peroxide (H_2O_2, 30%, Scharlau), PBS-Tween (containing 0.05% Tween 20, Sigma), 2-mercaptoethanol (ME, 99%, Acros organics), absolute ethanol (Scharlau) were used without any purification. The solutions were prepared in high purity de-ionised water obtained with a Milli-Q RG system (Millipore Ibérica, Madrid, Spain).

Synthetic oligonucleotides of the following sequences were purchased as lyophilised powder from Biomers (Germany):

- target amplicon LTA-α (63-mer sequence: 5′-GGG TTC CCT AAG GGT TGG ACT TCT CCC CAT GAC ACC ACC TGA ACG TCT CTT CCT CCC AAG GGT-3′
- thiolated DNA probe: 5′-SH-$(CH_2)_6$-ACC CTT GGG AGG AAG AGA CG-3′
- non-complementary target amplicon EXON16 (NTA) (62-mer sequence: 5′-GGG TTC CCT AAG GGT TGG ACC CTT ACC TGG AAT CTG GAA TCA GCC TCT TCT CTG ATG ACC CT-3′
- horseradish peroxidase (HRP) conjugated reporter probe at 5ĭof the sequence: 5′-HRP-TCC AAC CCT TAG GGA ACC C-3′.

Aliquots of 100 μM of stock solution of these oligonucleotides were prepared in water according to the supplier's instruction and stored frozen until needed. Working oligonucleotide solutions were prepared by appropriate dilution in hybridisation buffer.

2.2. Instrumentation

2.2.1. Electrochemical measurements

Cyclic voltammetry, potential step deposition of gold nanoparticles and chronoamperometric determination of target were carried out using an Autolab model PGSTAT 12 potentiostat/galvanostat controlled with the General Purpose Electrochemical System (GPES) software (Eco Chemie B.V., The Netherlands). A conventional three-electrode setup was used with the GC electrode modified with gold nanoparticles as the working electrode and a platinum wire as a counter electrode. An Ag/AgCl electrode (CHI 111, CH Instruments Inc., USA) served as the reference electrode. All potentials were reported with respect to this reference electrode. A magnetic stirrer provided the convective transport during potential step deposition and chronoamperometric experiments.

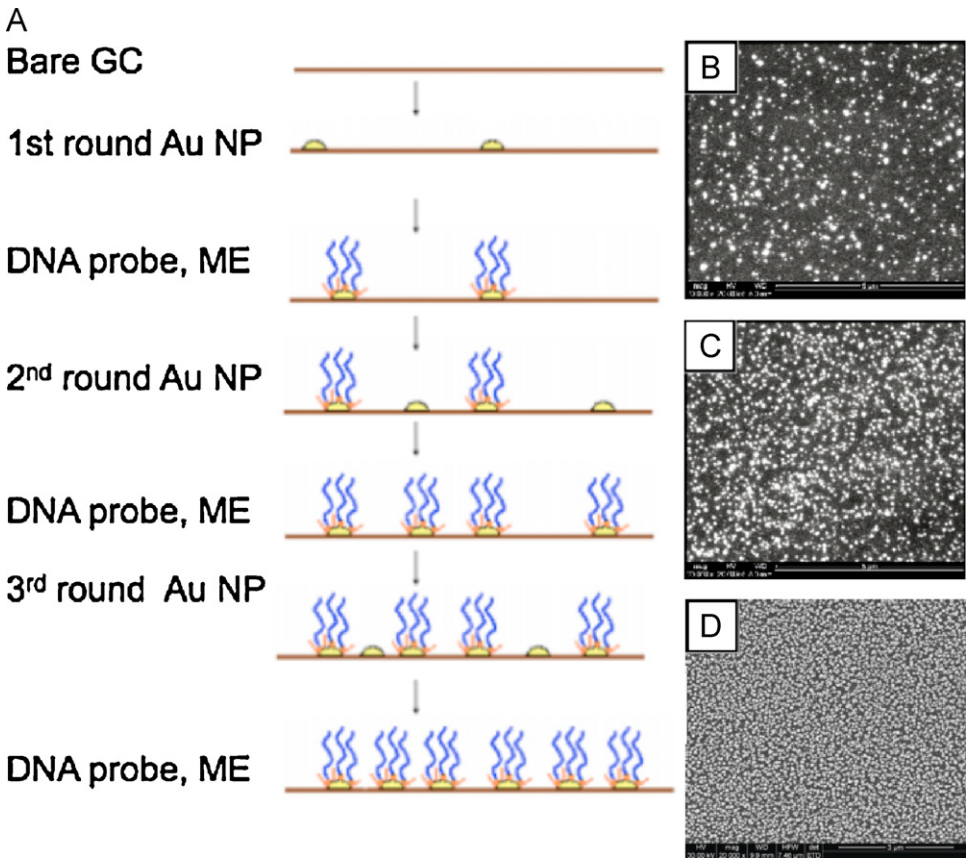

Fig. 2. (A) Illustration of the sequential electronucleation process for the detection of target DNA at electrochemically deposited gold nanoparticle using 1–3 rounds of electronucleation. Scanning electron microscopy images of gold nanoparticle modified GCEs after (B) a first round of electrochemical deposition and (C) three rounds of electrochemical deposition with protecting SAM of DNA capture probes (scale bars represent 5 μm). (D) Au-coated GCE surface following an electroplating treatment in 2 mM $KAuCl_4$ in 0.5 M H_2SO_4 for 30 s.

2.2.2. Scanning electron microscopy

Scanning electron microscopy for characterisation of the electronucleated gold nanoparticles on glassy carbon electrodes was carried out using a Fei Quanta 600 (USA) environmental scanning electron microscope at an acceleration voltage of 20 kV in a high vacuum mode and at a working distance of 5–10 mm. For nanoparticles number and size determination five SEM images were taken under the same magnification scale and analysed using image analysis software (ImageJ, National Institute of Health, v.1.38x)

2.3. Electrode cleaning and conditioning

Glassy carbon rods (Sigradur®, HTW Hochtemperatur Werkstoffe, Germany) with a length of 7 cm and a diameter of 3 mm were pressed into two layers of heat shrinking polyolefine tubes. One end of the rod, which serves as the electrode surface was polished with 0.3 μm alumina slurry (Buehler, USA). The electrodes were washed in water and sonicated in 50:50 (V/V) water:ethanol solution for about 15 min. After sonication, the electrodes were washed with water and electrochemically conditioned by potential scanning from 0 V to 1.4 V in 1 M $NaClO_4$ for at least five complete scans at 50 mV s^{-1} where the high background current due to glassy carbon oxidation diminished and a reproducible cyclic voltammogram was obtained. Afterwards the background current of the bare electrode was measured by cyclic voltammetry within the potential window of 0–0.7 V. The electrodes were used immediately following the cleaning and conditioning steps.

2.4. Electrochemical deposition of gold nanoparticles

Gold nanoparticles were deposited on polished and cleaned glassy carbon electrode by electrochemical reduction of potassium tetrachloroaurate (III) ($KAuCl_4$, Aldrich). A gold deposition bath was prepared by addition of 10 μL of 0.05 M $KAuCl_4$ (in 0.5 M H_2SO_4) to 5 mL of 0.5 M sulphuric acid (H_2SO_4). Subsequent nucleation steps were carried in a solution consisting of 10 μL of the 0.05 M $KAuCl_4$ added to 5 mL of 0.2 M potassium nitrate (KNO_3) so as to prevent the exposure of the DNA probes to low pH conditions. The gold nanoparticles were deposited by chronoamperometry where a pre-treatment potential of 1.1 V was applied for 5 s, followed by a reduction potential of 0 V applied for 5 s in a stirred gold deposition bath under stirring conditions. Plain gold electrodes were also prepared by electrochemical deposition of gold on GCEs by extending the deposition time to 30 s in 2 mM $KAuCl_4$ prepared in 0.5 M H_2SO_4.

2.5. DNA immobilisation and surface passivation

A working DNA probe solution was prepared at a concentration of 1 μM in 1 M KH_2PO_4, pH 3.8. For immobilisation, 20 μL of the solution was deposited onto the electrodes immediately after the gold electrodeposition step for at least 3 h and thoroughly washed in a stirred solution of PBS-Tween for 20 min in order to remove any weakly bound thiolated probes and minimise non-specific adsorption. The electrodes were finally rinsed in Milli-Q water for 10 min. The remaining uncovered gold was backfilled by immersing the prepared electrode in a 1 mM solution of 2-mecarptoethanol pre-

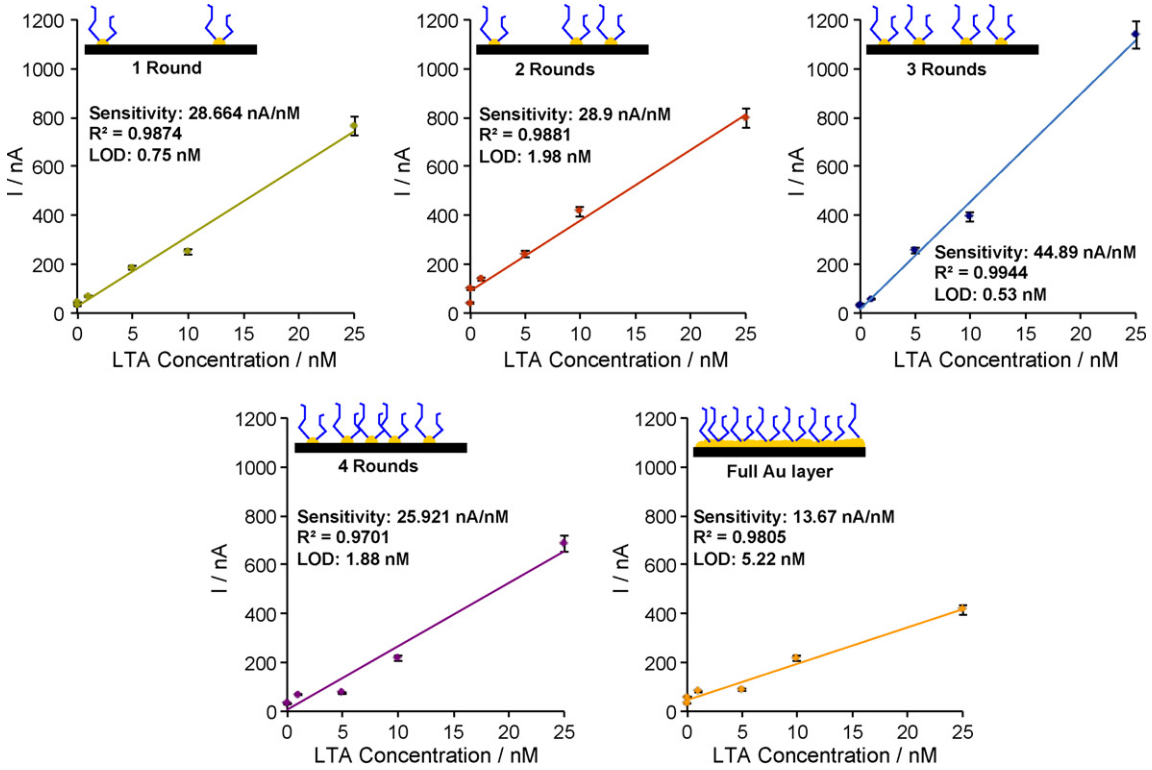

Fig. 3. Calibration curves for the detection of LTA in the range 0.1–25 nM for electrodes prepared following 1, 2, 3, or 4 sequential deposition/protection rounds of gold NPs as well as for a full gold layer electrodeposited.

pared in water for 30 min at room temperature. The electrodes were then washed in stirred solution of PBS-Tween 0.05% for 20 min and Milli-Q water for 10 min.

2.6. Hybridisation protocol

For hybridisation of the targets to the surface immobilised probes, 10 mM PBS buffer containing 600 mM NaCl, pH 7.4 (PBS-600) was used as hybridisation buffer. Oligonucleotide solutions in the concentration range 0.1–25 nM were prepared by appropriate dilution of the 100 μM target amplicon stock solution in hybridisation buffer. Following hybridisation for 60 min, the electrodes were washed in stirred PBS-Tween for 10 min and 20 μL of 10 nM reporter probe solution prepared in hybridisation buffer was deposited on the electrodes and left to interact with the surface duplex for 20 min. The electrodes were finally washed in stirred PBS-Tween for 10 min and rinsed thoroughly in hybridisation buffer before being transferred to the electrochemical cell.

2.7. Chronoamperometric detection of the hybridised target

Chronoamperometric detection was performed in 4.6 mL of stirred hybridisation buffer, at an applied potential of −0.1 V. After the signal stabilised (typically within 25 s), 200 μL of 0.5 M hydroquinone was injected followed by the injection of 200 μL of 0.5 M H_2O_2. The current required to reduce the hydroquinone oxidised during the regeneration of the HRP label (Fig. 1) was measured and used for the realisation of calibration curves (Harwood and Pouton, 1996; Sarma et al., 2009). Following chronoamperometric measurement, the electrodes were regenerated for repeated use by washing in stirred hot water (90 °C) for 3 min.

3. Results and discussion

3.1. Sequential deposition of gold nanoparticles for DNA biosensor applications

Gold nanoparticles were electrochemically deposited by reduction of the $KAuCl_4$ solution at the electrode surface. As we previously reported, the sequential deposition can be used to increase the particle number density of electronucleated gold nanoparticles (Soreta et al., 2008). In order to control the growth of the gold nanoparticles and ensure that new nanoparticles are formed during each nucleation round, freshly nucleated gold nanoparticles should be insulated with a capping agent. Efficient insulation guarantees that the growth of new nanoparticles during the next nucleation round only occurs at the glassy carbon surface by ensuring that previously deposited nanoparticles cannot act as nucleation centers. Blocking was achieved by immediately exposing the freshly prepared nanoparticles to a solution of thiolated DNA probe, followed by backfilling of the possibly remaining uncovered gold with a short alkanethiol such as ME. Fig. 2A. illustrates the strategy employed.

For the first deposition stage, the deposition bath consisted of 0.1 mM $KAuCl_4$ in 0.5 M H_2SO_4 as reported by Finot et al. (1999) and Soreta et al. (2008). However, in subsequent deposition stages, the supporting electrolyte was changed to KNO_3 so that the immobilised DNA would not be damaged by the low pH condition, using 0.1 mM $KAuCl_4$ in 0.2 M KNO_3 in all subsequent electronucleation rounds.

The prepared electrodes were inspected using SEM (Fig. 2B–D), which confirmed that the aggregation of gold nanoparticles did not occur during deposition. These observations corroborated evidence that the mixed SAMs of DNA and short chain alkanethiol provided sufficient insulation to enforce the creation of

new nucleation sites only at the GCE surface. SEM imaging also allowed the estimation of the density number of the nanoparticles to be approximately 14 nanoparticles $\times \mu m^{-2}$ (average particle size = 77.07 ± 2.03 nm, $n = 670$), and 23 nanoparticles $\times \mu m^{-2}$ (average particle size = 80.86 ± 4.16 nm, $n = 1097$) following 1 and 3 rounds of deposition/protection stages respectively (Fig. 2B and C).

3.2. DNA detection on sequential electronucleated gold nanoparticle modified GCEs

The performances of GCEs prepared following the electrodeposition of nanoparticles and planar gold thin films were assessed for the electrochemical quantification of DNA in the concentration range 0.1–25 nM (Fig. 3). The planar gold layer electrode exhibited the lowest sensitivity, i.e. 13.67 ± 1.74 nA $\times$ nM^{-1}, and the highest limits of detection (LOD), i.e. 5.22 nM, whilst electrodes prepared following either 1, 2, 3 or 4 rounds of electronucleation performed considerably better, as can be seen in Fig. 3. As the sequential depositions progress from 1 to 4 rounds, the LOD and sensitivity reached their optimal values of 0.53 nM and 44.89 nA $\times$ nM^{-1}, respectively after 3 rounds. The fourth deposition round did not further improve the performances of the electrodes suggesting that the electrode surface becomes highly covered by nanoparticles and DNA probes, limiting the efficiency of the hybridisation due to steric hindrance, as schematically depicted in Figs. 2 and 3. Eventually, as the number of deposition cycles is further increased, a situation where the GCEs are fully covered by nanoparticles that are no longer well isolated and insulated from each other can be envisaged.

3.3. Cross-reactivity

A non-complementary target at concentrations of 10, 50, and 100 nM was analysed on a thiolated capture probe monolayer formed after one round of gold nanoparticles deposition. The non-complementary control target sequence possessed a short section at the 5′ end that could match with the HRP-conjugated reporter probe used but that was otherwise non-complementary to the immobilised capture probe. All the concentrations tested gave a similar amperometric signal (20.33 ± 0.56 nA), equivalent to the blank measurement for this electronucleation stage, demonstrating that the non-complementary amplicon has no cross reactivity with the electronucleated immobilised capture probes and did not interact non specifically with the unprotected GCEs.

4. Conclusion

Nanostructured surfaces exploiting sequential deposition of gold nanoparticles were prepared as substrates for electrochemical genosensors, exhibiting better analytical performance than a planar gold electrode, with the highest signal enhancement observed after three rounds of nanoparticle deposition. The signal enhancement was attributed to the improved conditions for effective surface hybridisation between the capture probe and the target as a result of surface nanostructuring, as well as to the enhanced reactivity of the nanoparticles allowing a higher level of DNA probe immobilisation and higher catalytic properties of the deposited nanoparticles. Ongoing work is looking at the applicability of the approach to multiplexing on a single electrode surface, immobilising different probes after each round of deposition, with the reporter probe being linked to diverse enzyme labels, further demonstrating the potential of the strategy of sequential nanoparticle deposition and capping.

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